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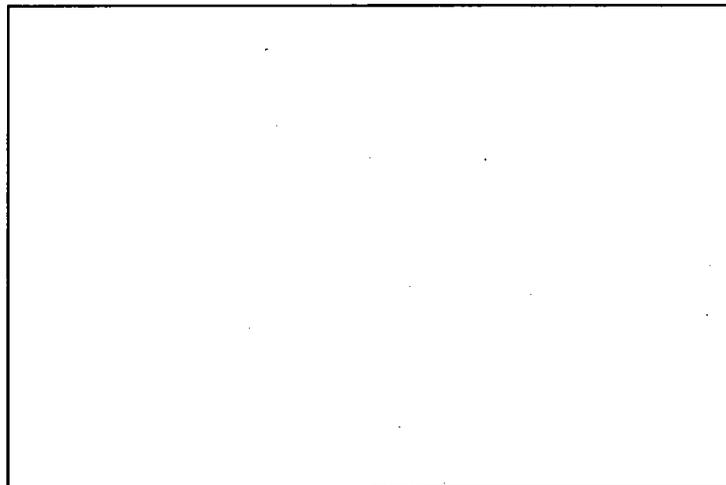
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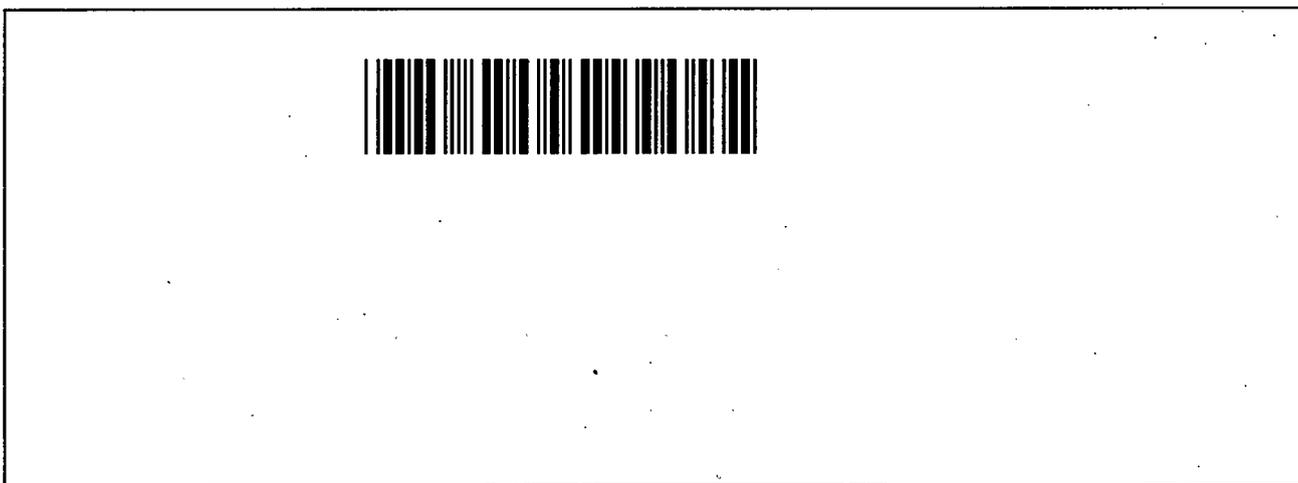
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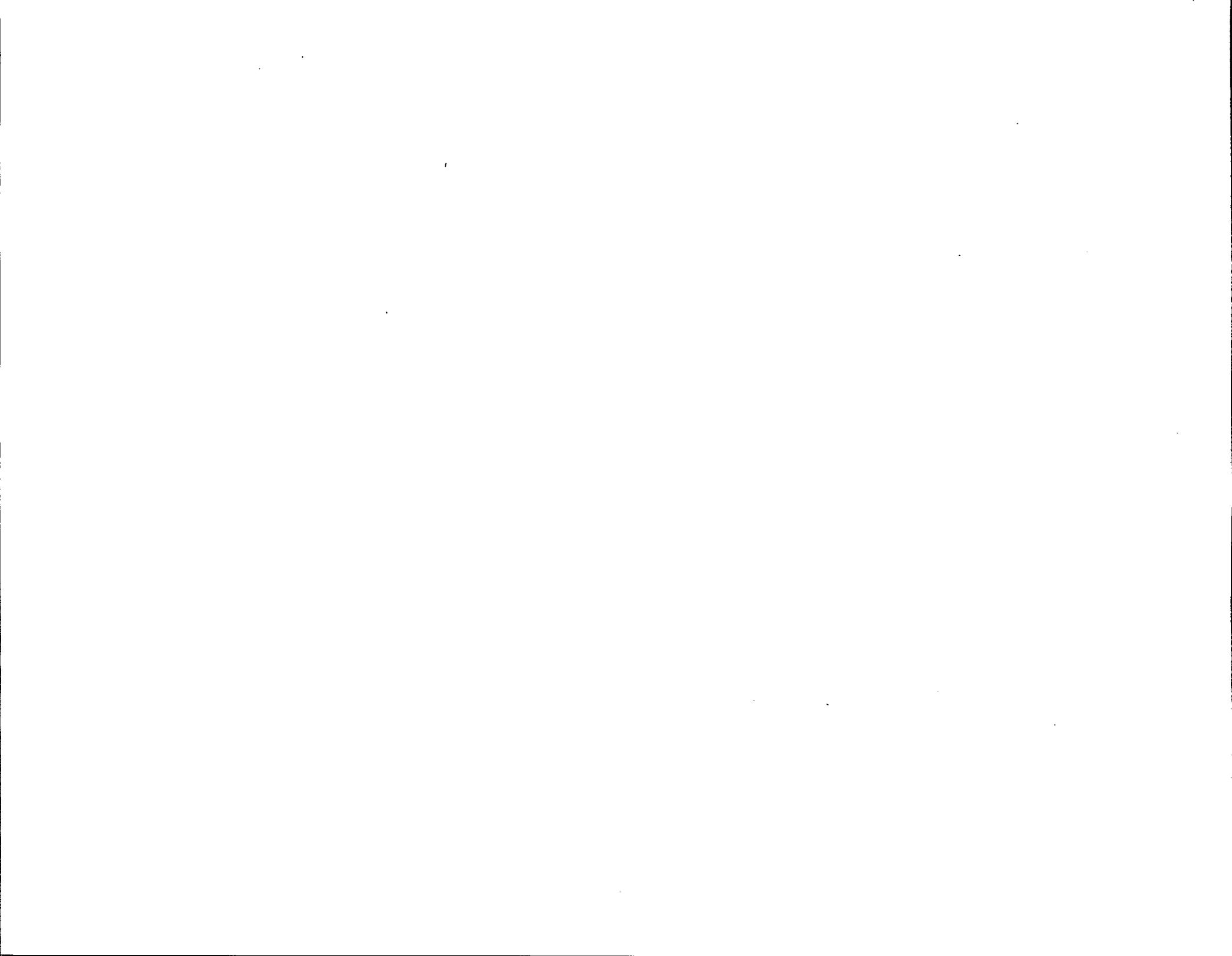
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Future Trends in the Assessment of Hazards from

Fission Product Releases

by

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ABSTRACTED IN NSA

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## 1. INTRODUCTION

In comparing and selecting sites for reactors from the point of view of safety, one considers the remote possibility of an accidental release of moderately large amounts of fission products and its effects in relation to the present and future distribution of population in the neighbourhood. At present until experience is gained of the reliability and safety of reactors, there is a tendency to site them remotely from centres of industry and population, although for economic reasons there will be a need to site large power reactors more closely to such centres in future. With, among other objectives, the aim of adopting in proper course of time less restrictive siting criteria, improvements are continually made in the intrinsic safety of reactor systems, and more sophisticated forms of reactor containment are devised, in order to reduce the possibility and scale of any fission product release.

Changes and improvements in reactor systems could affect the nature and proportion of an accidental release of fission products if this should occur in future. It is appropriate to consider what such a release and its radiobiological effects might be.

## 2. PREVIOUS ASSESSMENTS

Previous assessments have taken little account of special reactor or containment features, but have given a general appraisal based on release data, principally for metal fuels, and general meteorological and radiobiological information. The U.S.A.E.C. report WASH 740 is well known as are the illustrative calculations provided with the U.S.A.E.C. guide rules for reactor site selection. The latter assumed the following proportions of release - 100% noble gases, 50% volatiles, and 1% solid fission products. These figures are generally consistent with the experimental data from G. W. Parker et al at Oakridge. It has since been commented by Parker that while these assumptions still seem justified, they are extrapolations from experiments with small isolated specimens and take no account of "plate-out" or deposition on other fuel elements and reactor structures. In compiling a previous assessment, (U.K.A.E.A. Report A.H.S.B.(S) R.64 "An Assessment of Environmental Hazards from Fission Product Releases") I used the following proportions of release, which were derived from the observed release from the Windscale accident in 1957 - 25% iodine, 15% tellurium and caesium, and 0.1% strontium. It may be that the release from an air-cooled open circuit reactor as at Windscale has less relevance to present-day reactors. However it is interesting to note that the release from the SL1 accident which involved a quite different reactor type was again a preferential release of iodine with correspondingly lesser proportions of caesium and other isotopes. In both accidents the effects of containment or filtration were partial and future assessment should make allowances for some such effects.

It is useful to recall briefly to mind the principal hazards of fission product releases as outlined in previous assessments. The following figures are taken from report A.H.S.B.(S) R.64. A release from ground level in inversion conditions is considered. As is now well known, the dominant hazard would arise from the inhalation of iodine isotopes, particularly by young children. Taking as an example the release of  $10^3$  curies of iodine 131 together with other fission products in proportion, the maximum permissible emergency dose to the child thyroid (25 rem in the U.K.) would occur at 3 to

4 miles from the point of release. Contamination of cows' milk with iodine would be significant as far as 30 to 40 miles from the point of release; other effects such as contamination of crops and vegetables would probably be limited to an area within a mile from the point of release. Hazards from external beta-gamma radiation from the cloud of released activity and from ground deposition were also considered; the maximum permissible emergency dose (20 rad for children and pregnant women in the U.K.) would be received, if at all, at 200 to 400 yards from the point of release.

The hazard from an uncontained and unfiltered release is thus mainly from iodine, and realisation of this has led to considerable research and development on means of removal of iodine from gas discharges. The United Kingdom Atomic Energy Authority is carrying out an intensive programme of research and development on fission product filtration and many reactors are now equipped with absorber/filter systems as part of an overall containment system. As a result it is possible that if an accident occurred the release, if any, would be small and iodine might not be a significant hazard.

### 3. REACTOR FUELS AND FISSION PRODUCT RELEASES

The present nuclear power programmes in both France and Britain are based on the graphite-moderated, carbon dioxide cooled reactor fuelled with uranium metal clad in magnesium alloys. The release of fission products from uranium metal has been measured in several laboratories and the results are generally in accord with the figures mentioned earlier.

For many reactors there is a trend toward the use of ceramic fuels. The Advanced Gas-cooled Reactor, which is in operation at full power at Windscale, employs sintered uranium dioxide fuel canned in stainless steel. Oxide fuel is also used in a number of water reactors in the U.S.A. and elsewhere. During normal operations a significant release of gaseous and volatile fission products occurs by recoil, diffusion and other processes from the oxide and these accumulate within the fuel can. A small fraction of gaseous and volatile fission products would be available for release from the fuel can should this be ruptured as the result of an accident. However it is a characteristic feature of gas-cooled reactors, conferred by the thermal inertia of the massive graphite moderator, that fuel temperatures are kept within reasonable bounds even in quite severe postulated accident conditions such as sudden depressurisation. Taken together with the inert nature of oxide fuel with steel cans, this means that a release of fission products is unlikely.

Nevertheless it is customary, in order to assess possible consequences, to postulate that some overheating of fuel, possibly in the presence of air, has occurred. Experiments, in which the release of fission products was measured from oxide fuel overheating in this way, have been carried out by G. W. Parker at Oakridge and R. D. Collins at Windscale. These show quantitative release of gaseous and volatile fission products and only small quantities of solid fission products. One concludes that the relative proportions of fission products in a release from oxide fuel would not differ greatly from those expected in the case of metal fuel (although the likelihood of a release occurring would be much reduced in the case of a gas-cooled reactor). For water reactors there is a possibility of melt-out

if one postulates a sudden accidental depressurisation with additionally failure of the emergency core spray. Parker at Oakridge has measured the release of fission products from uranium dioxide on melting. Release of gaseous and volatile fission products was nearly complete but the release of solid fission products was small. Hence our previous general conclusions as to the relative proportions of fission products in the release can be applied to the case of oxide fuels.

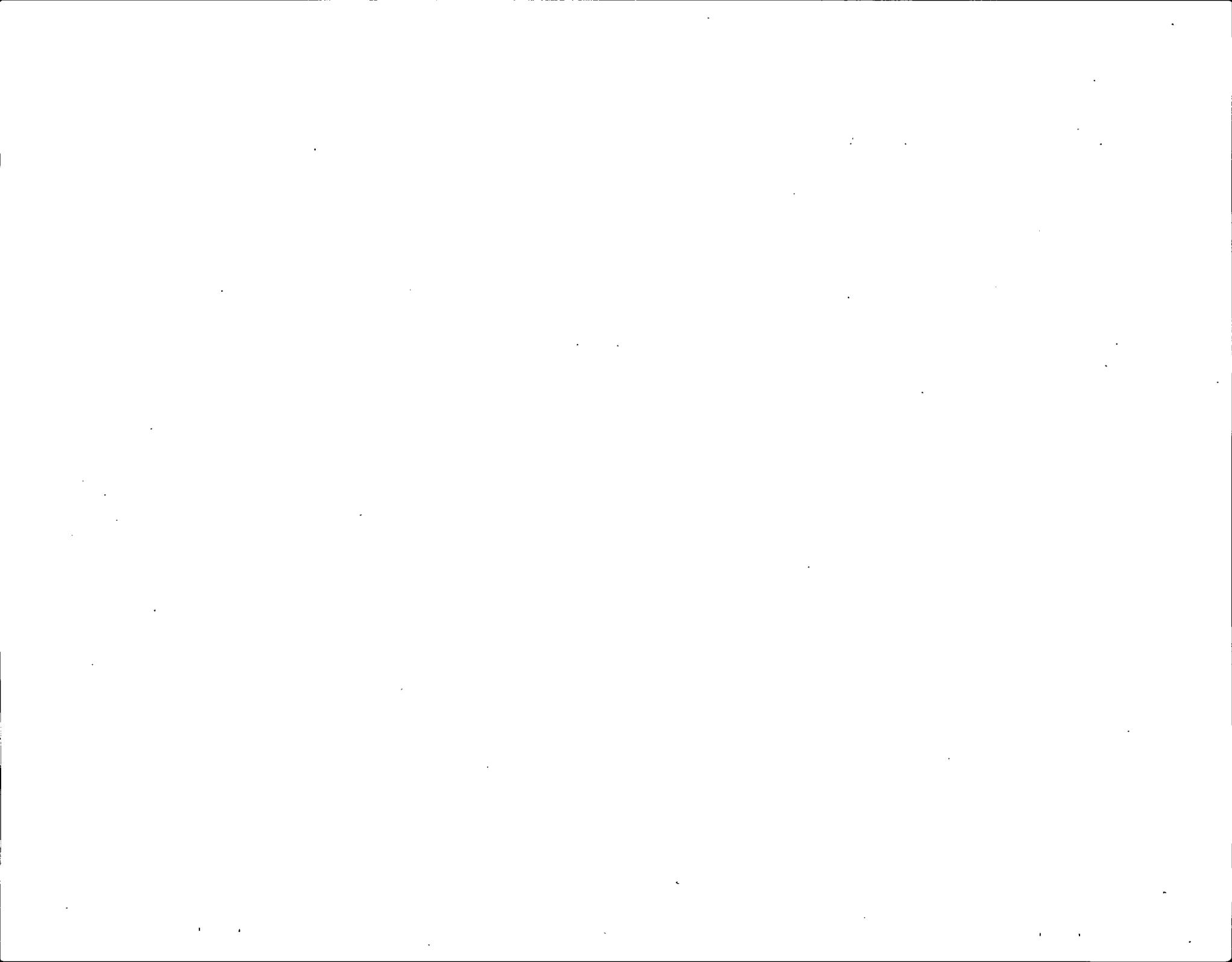
Uranium carbide is being developed as a reactor fuel. Sintered or fused carbides, which are usually canned in stainless steel, are usually superior to oxide in fission product retention at operating temperatures. In accident conditions their behaviour might be less favourable since they react readily with air and steam. Coated particle fuels which use carbide particles embedded in a graphite matrix are being developed for high temperature gas-cooled reactors such as the O.E.C.D. DRAGON reactor at Winfrith Heath. From preliminary results it seems likely that coatings can be developed which will limit the release of fission products, at operating temperatures to a small quantity of noble gases. Because of the all-ceramic form of these fuels, the release in accident conditions may well be small provided air can be excluded.

#### 4. REMOVAL OF IODINE BY DEPOSITION AND OTHER PROCESSES

For an unrestricted release of fission products from reactor fuel as the result of an accident, the foregoing confirms that iodine-131 would constitute the principal health hazard. It is important to consider how deposition and other processes in the reactor and its containment would affect this conclusion.

Iodine mass is generated in a reactor at a rate of about 5 milligrams per megawatt-day, and calculations for typical cases show that initial concentrations in the range 1 microgram/metre<sup>3</sup> to 100 milligram/metre<sup>3</sup> could be formed. This is well below the saturation vapour pressure of molecular iodine at normal temperatures, but deposition can occur by chemical and physical absorption. Some iodine will be adsorbed on the graphite of gas-cooled reactors, and calculations, based on experimental determinations of iodine vapour pressure over specimens of reactor graphite, show that between 50 and 80% of iodine might be retained, depending on temperature. In a water reactor accident some iodine would be retained with the reactor water, the relative proportions in the liquid and gaseous phase depending upon temperature, volume ratio of the two phases, and initial concentration. In typical cases the proportion retained by the water would be anything from 10% to 90%. The behaviour of sodium-cooled reactors appears particularly favourable. Atomics International have found, from laboratory experiment and from a reactor incident, that iodine and other fission products, with the exception of the noble gases, are almost completely absorbed in passage through sodium. From the above it appears that some retention of iodine by moderator or coolant can be expected, and that this effect would be marked in a sodium-cooled reactor, unless, of course, the sodium itself were dispersed.

Dry deposition of iodine upon containment surfaces is a powerful mechanism for removal of airborne iodine. This was demonstrated for low



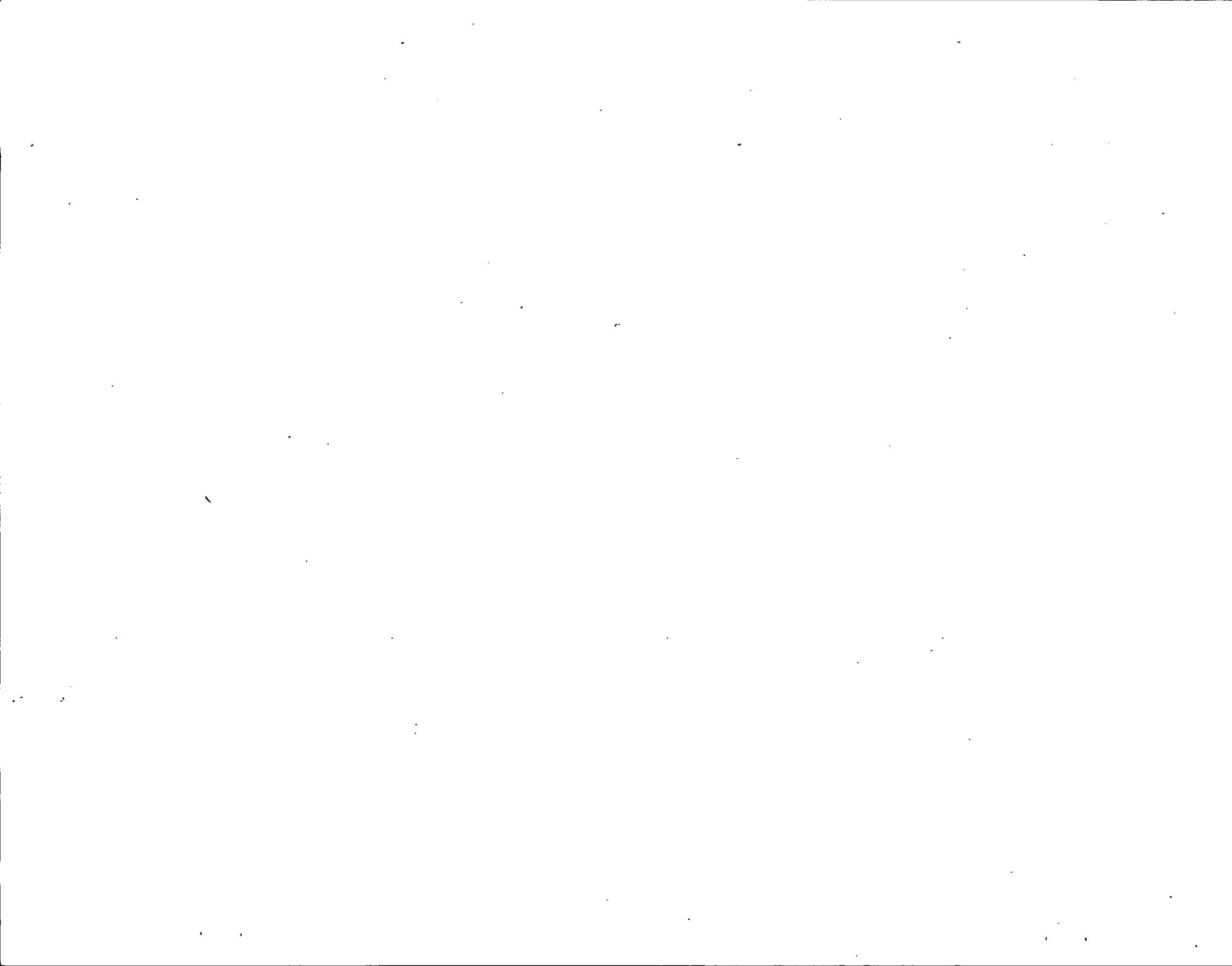
concentrations of iodine in the well-known experiments carried out by Chamberlain and his co-workers, in which they released iodine in the containment shells of the DIDO and PLUTO reactors at Harwell. Experiments were later performed by Croft at Winfrith Heath, in which sufficient iodine was released in a specially prepared room to give initial concentrations as high as are ever likely to occur in a reactor accident (about 100 milligram/metre<sup>3</sup>). Croft's measurements, which were extended over many hours and days, showed that the deposition of iodine continued until airborne activity was reduced by several factors of ten. The decline of airborne activity was approximately exponential with a half-life for deposition equal to  $0.693 \frac{V}{s}$  where  $s$  is the surface area per unit volume of the containment space and  $V$  is the deposition velocity of the activity. Extrapolating to full-scale reactor containment one finds the half-life for deposition would be in the range  $\frac{1}{2}$  to 10 hours. The total leakage from a pressurised containment is simply  $LI/T$  where  $L$  is the leak-rate,  $I$  the initial airborne activity and  $T$  the half-life for removal. This means that most of the leakage of activity takes place during the first deposition half-life and a substantial reduction in total leakage results from making this as short as possible.

Although in Croft's experiments there was no real evidence of surface saturation it may be that there was some limiting value of concentration in air, perhaps, a few micrograms/metre<sup>3</sup>, below which deposition practically ceased. This limit may have been related to the presence of iodine in other than molecular form. One recalls Chamberlain's observation that some iodine becomes attached to condensation nuclei in the atmosphere and also Eggleton's recent identification of methyl iodide and other compounds as constituents of iodine aerosols. Croft also found evidence of desorption of iodine from samples removed from the contaminated atmosphere for a period of several days. Therefore even though much iodine would be removed by deposition where a reactor is enclosed by a vessel, shell, or building, a need for additional clean-up by some form of filtration or absorption system will remain.

##### 5. REMOVAL OF IODINE BY FILTRATION/ABSORPTION PLANT

It would be inappropriate in this paper to attempt to describe the many forms of filtration or fission product absorption system which have been proposed and developed in recent years in response to the need for means of controlling fission product discharges. The one which is most commonly used in the United Kingdom and also elsewhere consists of a series combination of coarse and "absolute" particle filters with a bed of activated charcoal granules. This system is found to give, when properly designed installed and used, a decontamination factor (ratio of concentrations in air at inlet and outlet) exceeding  $10^4$  for iodine, and similar figures for most particulate fission products. (As yet one is not completely reassured about the efficiency of "absolute filters" against fission product strontium, by the few measurements which have so far been reported. However the reported fractional release of this element is usually small and this may outweigh the possible reduction in filtration efficiency, if confirmed).

One use of such a system is as a recirculation clean-up plant to treat gases in the reactor vessel or containment prior to leakage or discharge. Simple mathematics show that the effect of such a system is to reduce the airborne activity exponentially. The half-life for this process is  $0.693 T/F$  where  $T$  is the time required to pass the total gas volume once through the plant and  $F$  is its once-through filtration efficiency



expressed as a fraction. From engineering and economic considerations this time usually turns out to be of the order 1 - 4 hours. In simple terms a recirculatory clean-up system enhances the effect of deposition in the containment and provides a substance and site for more permanent retention of fission products.

Methods of fission product removal which involve the rapid injection of absorbing substances into the containment offer the possibility of reducing the half-life for deposition to a few minutes. Griffiths of Safeguards Division, U.K.A.E.A. has recently discussed the theoretical and practical limits to the usefulness of sprays in removing iodine. Silverman at Harvard has experimented with foam injection for removing iodine and particulates, and a few years ago Rosinski at Armour Research investigated the possibilities of injecting chemical scavengers to bring down particulates. To date, apart from spray systems in some water reactor containments, these have not generally been used in reactor engineering designs. A pool suppression system is used in some recent reactor containments. This has as its primary purpose that reactor steam from a circuit rupture should be condensed by injection into a pool of water in order to reduce or eliminate containment pressure, but rapid removal of most fission products in the pool appears to be a useful by-product.

Because of its proved high efficiency the conventional plant consisting of filters and charcoal bed may be used on a once-through basis to clean up a direct discharge to the atmosphere of reactor and containment gases. The discharge, which should preferably be made from a high stack, can be used to create a sub-atmospheric pressure in the containment and so eliminate active leakage at ground level. Many different containment systems have now been constructed, some with single and others with double shells, some with recirculatory clean-up others with once-through filtration and many with both. It would scarcely be appropriate to attempt to discuss the performance of these systems in detail here, but it would be useful to consider generally how such containment might affect the nature and consequences of a fission product release if this should occur.

The principal effect, of course, should be a substantial reduction in the size of the release. Taking 500 MW thermal power as typical of present-day large power reactors, individual isotopes of importance as health hazards, such as iodine 131, caesium 137, and strontium 90, would be generated in the fuel to the amount of  $10^7$  curies each (in very round numbers). The tests and experiments carried out so far illustrate separate aspects of the problem and their solutions, and suggest the following assumptions. Release from the fuel would in all probability only be partial. Deposition in and around the reactor and containment might provide several factors of  $10^3$  removal. The containment leak-rate, if this were relevant, might be of the order of 1% per day. A clean-up plant, if a controlled discharge were made, might give a decontamination factor of order  $10^4$ . If these factors can with certainty be assumed, the total release could be held to the range 1 -  $10^2$  curies and in some cases even less. For release from a stack, acceptable emergency releases of the previously mentioned isotopes probably fall in the range 10-100 curies, if based on inhalation hazard, or 0.1 to 1 curies, if based on ingestion of cows' milk. It seems then that a solution to the problem of fission product release is in sight. But much more exacting proofs than are at present available will rightly be required before evidence of this type becomes fully acceptable to the various national reactor safety authorities. To this end elaborate and realistic trials are under preparation.

## 6. HAZARDS FROM A RELEASE OF FISSION PRODUCT NOBLE GASES

As yet no practicable method exists for removing fission product noble gases from the large quantities of gas or steam which might be released in a reactor accident. (Liquid nitrogen-cooled charcoal delay beds are, of course, a practical means of rare gas clean-up where the gas flow is small). The possibility of discharging noble gases to atmosphere in an accident raises problems of external radiation dose from the plume and of the inhalation or ingestion hazards which might arise from solid daughter products. Calculations can be made of these effects, and, as is customary, the figures which follow apply to fairly adverse conditions with a low wind speed.

It is convenient to consider a 100% release of fission product noble gases from an amount of fuel corresponding to 4 megawatts thermal power. (It is of incidental interest that this order of power corresponds to the rating of a single fuel channel in many present-day advanced reactors). From an operating reactor this release would involve initially  $10^6$  curie-MeV, approximately, of noble gases. The external dose of gamma radiation delivered to a person standing 500-1000 metres downwind under the plume would be about 25 rad where the release was from a 15 metre high stack, and say 10 rad for release from a 50 metre stack. Evidently there is a quite restrictive limit to the noble gas activity which can safely be discharged in this way. A useful advantage is gained by imposing a delay between emission of the gases and their release to atmosphere, by the interposition of one or more containment spaces between the reactor and the point of discharge. In an ideal case radioactive decay within the containment for about 6 hours would reduce the dose subsequently received from the cloud by a factor of 10, and 4 days delay would confer a factor of 100. The advantages accruing from delay volumes in containment systems were discussed recently by O'Neil and Edwards.

Daughter products of the noble gases which are of interest are listed in Table I. Rubidium-88 and caesium-138 are both energetic beta-emitters. Since they would be deposited by diffusion and impaction on objects immersed in the plume they might become apparent as short-lived skin and clothing contamination after the passage of the cloud. Such contamination could be quite high - possibly  $1 \mu\text{c}/\text{cm}^2$  for the release previously considered; the integrated beta dose to the skin would be about 10 rem, and the gamma dose about 1 rem.

Of the three long-lived daughters listed in Table I, strontium-89 would be the greatest hazard. Up to 5 curies of strontium-89 would be dispersed by the release previously considered, but only 5 millicuries of strontium-90 and 40 millicuries of caesium-137. Even a short delay before release could greatly reduce the proportion of strontium-90 released outside the containment. The quoted release of 5 curies strontium-89, if from a low level, might just be enough to invoke a ban on milk consumption about half a mile from the source. There would be no significant inhalation hazard from strontium-89 unless the release, if instantaneous, involved at least 100 MW of fuel. Any appreciable delay in the release of the parents would practically eliminate the daughters from the discharge, but even if there were no delay the external gamma radiation from the cloud of gases would be more important than ingestion or inhalation of the daughter products.

## 7. CONCLUSION

There are good prospects that as a result of an improved intrinsic safety of reactors and the development of reactor containment and fission product clean-up systems, the release from a future reactor accident, if one should occur, would be small. The escape of iodine in particular should be effectively prevented by these measures. However certain doubts still need to be resolved, for example, on desorption of iodine and on the trapping of gaseous iodine compounds. Some further assurance is also needed on the filtration of strontium. Many doubts will be removed if systems are tested under conditions closely resembling those they are designed to meet - that is to say, against fission products released from overheated fuel on a realistic scale and in realistic conditions of gas composition, flow and temperature. A programme of experimental work along these lines is at present being carried out by the U.K.A.E.A. at its Windscale Laboratories.

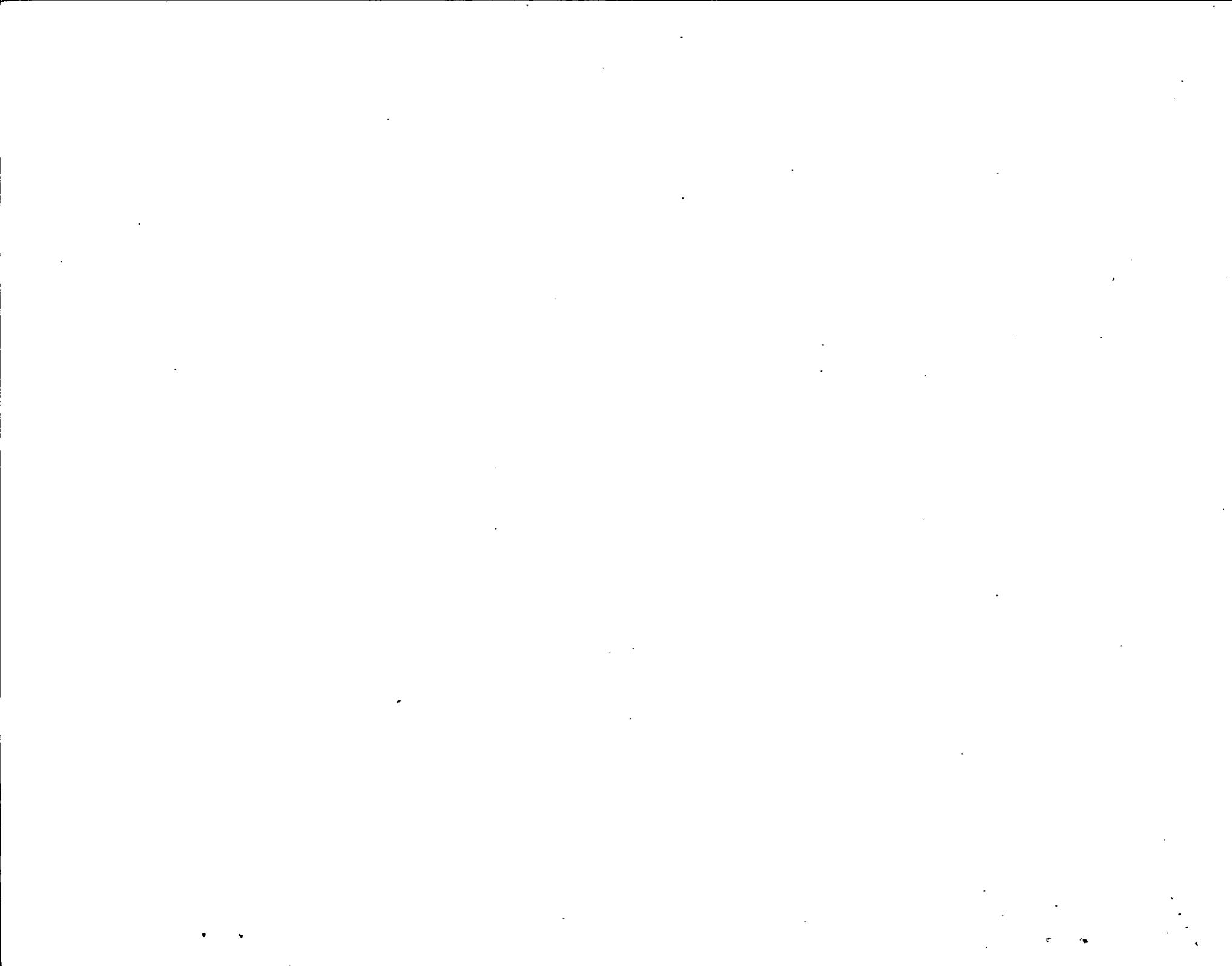
For some containment systems the ultimate hazard in an accident might be from the discharge of fission product noble gases. External gamma radiation from the cloud of gases would be the most important effect and would impose a quite restrictive limit on the noble gas activity which could permissibly be discharged. Hazards from solid daughters of noble gases are of secondary importance compared to this. These include a beta dose to the skin from rubidium-88 and caesium-138, and the possibility of some contamination of milk by strontium-89 for moderately large discharges. Useful safety factors can be achieved by controlling and delaying the discharge using one or more containment spaces and by directing any discharge to a high stack.

Ultimately one result of the development of methods of removing iodine and other volatile or particulate fission products should be that the potential hazard from these is no greater than that of the fission product noble gases. The total hazard will then be much diminished. This will be a trend which will require to be re-assessed from time to time. Taken together with improvements in the intrinsic safety of reactors, this trend in the assessment of fission product hazards should justify the progressive relaxations in site selection criteria which may be required for future power reactors.

TABLE I

SOME DAUGHTER PRODUCTS OF FISSION PRODUCT NOBLE GASES

Parent	Daughter of Interest	Remarks
Kr 88 (2.8 hr)	Rb 88 (18 m)	decays to stable Sr
Kr 89 (3.2 m)	Sr 89 (50 d)	via Rb 89 (15 m)
Kr 90 (33 s)	Sr 90 (28 yr)	via Rb 90 (2.9 m)
Xe 137 (3.9 m)	Cs 137 (30 yr)	decays to stable Ba
Xe 138 (17 m)	Cs 138 (32 m)	decays to stable Ba



## REFERENCES

1. "Theoretical possibilities and consequences of major accidents in large nuclear power plants". U.S.A.E.C. Report WASH-740, March, 1957.
2. Beattie, J. R. "An Assessment of environmental hazards from fission product releases" U.K.A.E.A. Report AHSB(S) R.64 (1963).
3. Chamberlain, A. C. et al. "Behaviour of iodine vapour in air" Disc. Faraday Soc., No. 30, pp. 162-169, (1960).
4. Megaw, W. J. and May, F. G. "The behaviour of iodine released in reactor containers" J. Nucl. En. parts A/B, vol. 16, pp.427-436, (Sept. 1962).
5. Morris J. B. et al "Removal of low concentrations of iodine from air on a plant scale". J. Nucl. En. parts A/B vol. 16 pp. 437-445, (Sept. 1962).
6. Eggleton, A.E.J. and Atkins, D. E. "Identification of radio-iodine compounds formed on releasing carrier-free iodine-131 in air". A.N.S. Transactions, vol. 6, No. 1 p. 129, (1963).
7. Croft, R. F. and Iles R. S. "The surface deposition behaviour of high airborne concentrations of iodine". U.K.A.E.A. report AEEW-R.265, (1963).
8. Griffiths, V. "The removal of iodine from the atmosphere by sprays" U.K.A.E.A. report AHSB(S) R.45 (1963).
9. O'Neil R. and Edwards A. R. "A review of containment schemes for water reactors" Proceedings of I.A.E.A. Symposium on Reactor Siting, Bombay, 1963, pp. 245-273.

